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AMERICIUM. ITS BEHAVIOR IN SOIL AND PLANT SYSTEMS

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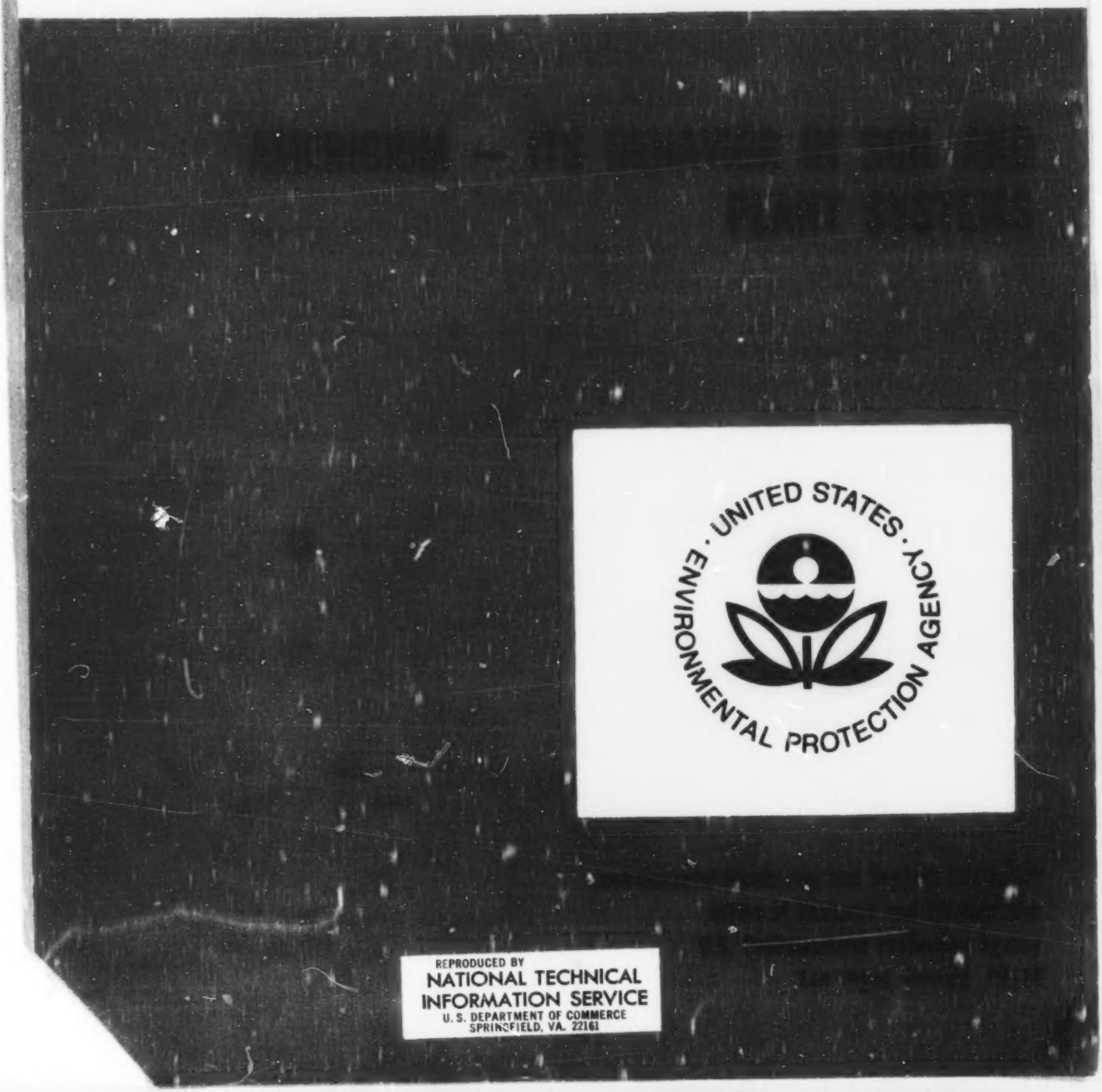

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The small amount of data available on the behavior of americium in plant and soil systems is reviewed and found lacking in several critical areas. Although some studies have been done on the physical and chemical interaction of americium on these systems, most of them were short-term and limited in scope. As americium is classified as a hazardous radionuclide and is likely to increase in importance as an environmental pollutant, further study is suggested. Also, the use of americium as a tool for measuring various soil parameters is discussed.

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INTRODUCTION

The release of long-lived radioactive elements into the biosphere in past years by atmospheric nuclear testing and currently by the expanding nuclear power industry has prompted the need for assessing the impact of these pollutants on our environment. Studies of various radionuclides by Hanson and Eberhardt (1971), Romney *et al.* (1971), Williams (1967), and Koranda *et al.* (1969), have shown that once released, many radioactive pollutants tend to accumulate in the environment in different forms and also in one of the many trophic levels of a biological system. Although little is known about the behavior and transfer kinetics of many radioactive pollutants in the soil and plant environment, the foregoing studies have shown that their movements in both the edaphic and biotic compartments were found to be, in general, very slow. As reported by Price (1973a), the final assessment of environmental impact will show that pollutants having long residence times will increase in relative importance with time, especially in areas where they are uncontained within the biosphere.

Some of the longest lived and abundant radioactive pollutants being released into our environment are the transuranic elements. For example, Jacobs and Gera (1969) have projected that the accumulation of plutonium and its decay products from the United States nuclear power industry will exceed 1,000 megacuries by the year 2020. Another investigator (McKay 1961) reported that the most biologically hazardous radioelements being produced by our nuclear industry in addition to plutonium are other actinides, americium, neptunium and curium. This was substantiated by Denham (1969) when he reported on the radiological hazards of the transuranic elements.

Three excellent review articles have been published on the chemical and physical behavior of the transuranic elements in biotic systems. Two of these authored by Price (1973a) and Francis (1973) are primarily concerned with terrestrial behavior. The other, authored by Noshkin (1972), is concerned with the behavior and dissemination of these radioactive pollutants in aquatic environments.

It is evident from the scientific literature that most biological investigations conducted concerning the transuranic elements were focused primarily on plutonium, which is considered to be the most dangerous to man. For the most part, the other transuranic elements were studied in an ancillary manner, and consequently, relatively few reports exist concerning their fate and behavior in the environment.

During the past few years, the scientific community has become increasingly aware of americium as being a major radioactive pollutant. Of its twelve isotopes, americium-241 is generally considered to present the most serious environmental hazard. Americium-241 enters the environment primarily as a decay product of plutonium-241, which is released into our biosphere as a by-product

of both the nuclear testing program and the nuclear power industry. Americium-241, like its parent, is a long-lived radionuclide that will accumulate in our environment and may become more biologically available with time. Poet and Martell (1972) estimated that the maximum environmental impact of americium-241 following an accidental release of plutonium-241 would probably occur in 70 to 80 years. Also, Major *et al.* (1974) suggested that high priority be given to investigations involving americium, as nearly 26 years have passed since the initial release of plutonium-241 into our environment. This review was undertaken to assemble and summarize the available data concerning the distribution and behavior of americium in both soil and plant systems and also to identify areas of needed investigation.

AMERICIUM IN THE SOIL SYSTEM

The main research goals of many investigations have been to determine the pathways and rates of radionuclide transfer among the many components of both native and agricultural ecosystems. Studies of soil, soil-plant, and plant systems can serve to show mechanisms by which americium could become incorporated into biological systems. Many studies have been initiated following accidental or continual releases of radioactive pollutants into our environment, while others have been conducted under controlled conditions using radioisotopes as tracers. Both types of investigations have added to our understanding and knowledge of the complexity of these pollutants in our environment.

Many studies involving transuranic elements in soils have been conducted on or near the U.S. Energy Research and Development Administration's (formerly the U.S. Atomic Energy Commission) Nevada Test Site following nuclear testing. Experimentation has also been conducted in areas used for the disposal of both liquid and solid radioactive wastes. These studies were initiated due to past practices of using soils to sorb and retain radionuclides.

Some of the earliest work conducted concerning actinide behavior in soils was accomplished by Rhoads (1957). He reported various characteristics of plutonium adsorption by soil collected near the Hanford Disposal Site. Approximately 10 years later, Hajek (1966), using soils collected from the radioactive waste cribs near Richland, Washington, reported on the mobility of americium in soil. After leaching aliquots of soil containing americium and plutonium with both water and a solution of 1*N* $NaNO_3$, Hajek found that 7.5% of the americium was removed from the soil with the water treatments while 33% was leached with the $NaNO_3$. A similar study using gravity-fed soil columns to investigate the ionic exchange properties of americium-241 was conducted by Knoll (1969). After applying various organic compounds tagged with americium-241 to soil columns, he reported that the soil had little or no effect on removing the americium from any of the compounds tested. However, americium previously applied to soil was rapidly and completely leached by both di-(2-ethylhexyl) phosphoric and hydroxyacetic acids.

A study by Wallace (1972a) showed that the common chelating agent diethylenetriaminepentaacetic acid (DTPA) can be used to increase the uptake of americium-241 by plants and that it was also an excellent soil extractant for americium-241. Cline (1968) revealed that soil pH affects the downward

migration rate of americium-241. He applied $^{241}Am(NO_3)_4$ to two different soils, one having a pH of 4.5 and the other 7.5. His results showed that, after each column was leached with 100 inches of irrigation water, in excess of 98% of the americium was retained in the top 1 centimeter of the acid soil, whereas only 76% remained in the top layer of the basic soil. Maximum depth of penetration was noted when americium was detected near 20 centimeters in the basic soil and only at the 5-centimeter depth in the acidic soil.

The foregoing studies concentrated on the extraction or leaching of americium from soil with a variety of solvents or solutions. From these findings, it is evident that americium interacts both chemically and physically with various types of soil. However, it should be noted that these studies were limited in scope and definitive deductions cannot be derived from the resulting data. This is primiarily due to the physical and chemical complexity of soils and their effects on the various chemical forms of americium, which apparently have not been extensively investigated relative to americium behavior.

The application and use of americium as a tracer in soil studies, such as in the determinations of both soil density and soil water content, have increased in recent years. King (1968) reported that the use of americium for these determinations was not fully investigated until after 1962 due to its limited availability. Since then, King (1968), DeSwart and Groenevelt (1971), Corey *et al.* (1971), Bridge and Collis-George (1973), and Reginato (1974) have all shown that americium-241 is a suitable gamma emitter for these determinations. Basically, the method involves measuring the attenuation of collimated beams of monoenergetic gamma rays passing through soil containing varying amounts of water. The two advantages of using americium-241 in preference to other tested gamma-emitting radionuclides for these measurements are: 1) it has a long half-life of 458 years, and 2) it has a relatively low-energy gamma ray, having a major peak at 60 keV, therefore requiring less shielding for operator safety.

The transfer, mobility, and biological availability of americium in soils may be enhanced by soil microorganisms. For example, Au (1974) indicated that plutonium was assimilated and taken up by *Aspergillus sp.* from a plutonium-contaminated malt agar. His work suggests that acid-producing soil molds may solubilize other actinides for assimilation and uptake by soil microorganisms and higher plants. Unfortunately, there does not appear to be any conclusive data in the literature concerning microbial effects on americium. However, studies are underway to determine the role of selected soil microorganisms in the complexing of various soil constitutents in soils contaminated with plutonium or americium.

AMERICIUM IN THE PLANT SYSTEM

Rediske *et al.* (1955) indicated that the majority of the radioactive nuclides incorporated into biological systems entered via plants. One means of incorporation is by the interception and retention of airborne contaminants by aerial portions of plants with subsequent transport between the different trophic levels. Another method is the deposition of contaminants on soils with eventual root absorption.

As with soil investigations, relatively few plant studies have been

conducted to determine the magnitude of root absorption using both contaminated soil and hydroponic rooting media. The data from these investigations indicate that the amount of americium absorption is similar to that of many heavy metals (Price, 1973a). Apparently, the only studies concerning biological incorporation of alpha-emitting nuclides by foliar deposition are being conducted at the U.S. Energy Research and Development Administration's Nevada Test Site. In these studies, Barth (1974) and Smith (1974) showed that the concentration of alpha-emitting nuclides increased in cattle rumen ingesta following the ingestion of *Eurotia lanata*. Their conclusion was that this increased concentration was due primarily to the relatively high deposition and physical retention of particles by this species. While studying plant uptake and the retention of selected actinides, Romney *et al.* (1974) also found this plant species to be an excellent collector of airborne contamination.

Foliar uptake and retention following the immersion of orange leaves in various solutions containing americium-241 were studied by Wallace *et al.* (1969). They found that the absorption of americium-241 increased with an increase in solution pH. However, when the chelating agent DTPA was added to the solutions, a decrease in americium-241 absorption occurred. The maximum absorption of americium with DTPA occurred at a solution pH of 2.0, decreased at a pH of 5.0, and leveled off at a pH of 8.0. They concluded that at a pH of 2.0 the DTPA had no effect presumably because the complexing of americium-241 cannot take place under highly acidic conditions.

The uptake of radioisotopes via the roots of plants generally results in a long-term radionuclide plant burden. For example, Romney *et al.* (1970) found that during a 5-year cropping study, a consistent yearly increase in plant tissue concentration occurred. They concluded that the increase in uptake was due either to an ever-expanding root system, which would physically increase the probability of the roots coming in contact with the radionuclide, or that a chemical transformation occurred as a result of an increase in soil organic matter from root tissue decay.

Several different chemicals were added to soils containing americium to determine their effects on plant uptake. This study by Hale and Wallace (1970) showed that a substantial increase in the amount of americium occurred in the aerial portion of bean plants grown in soils spiked with 10 microcuries of americium-241 (in HNO_3) following applications of two different chelating agents. The chelating agents DTPA and ethylenediamine di(o-hydroxyphenylacetate) (EDDHA), which were applied at a rate equivalent to 10 pounds of iron per acre, increased the root uptake of americium by nearly 1,000 and 500 times, respectively.

In similar experiments, Wallace (1972a, 1972b) grew soybeans and citrus plants in soils spiked with americium. His data showed that the incorporation of americium-241 in both species increased with applications of DTPA. The translocation and percentage composition of absorbed americium-241 in bush beans grown in a nutrient solution spiked with americium-241 was also investigated. His data showed that the majority of the americium was located in the leaves (62%), followed by the roots (25%), and the stems (13%). The data from a study to determine the influence of soil pH on the chelation of americium-241 by DTPA were also reported by Wallace. The data showed that the greatest uptake of americium by bush beans occurred in soils with a pH of 7.7. Also the

ratio of americium-241 in the leaf versus that in the stems was greatest at this pH.

The uptake of americium by beans grown in a standard Hoaglands solution (Hoagland and Arnon 1950) containing 0.9 μCi/liter of $^{241}Am(NO_3)_3$ and barley grown in two different soils spiked with 1.8 μCi/g of $^{241}Am(NO_3)_4$ was investigated by Cline (1968). He reported that the ratio of americium in the leaves to that in the rooting media, soil or solution, was nearly identical for each species. The pH of the soils used by Cline, one having a pH of 4.5, the other 7.5, did not appear to influence the uptake of americium. This is interesting as Wallace (1972a) reported that soil pH without the addition of chelating agents did affect the uptake of americium. His data showed that a greater amount of americium-241 was taken up by plants grown in soils at a pH of 5.3 than those grown in soils with a pH of 8.7.

Evidence of the toxicity of americium to plants was also reported by Cline (1968). He found that secondary roots were not formed by pea plants grown in a nutrient solution containing americium at a concentration of 0.1 microcurie per liter. After an identical treatment using plutonium-239, he concluded that plutonium was not as toxic to plants as americium and also that americium-241 could not be used as a reliable tracer for plutonium-239 due to a difference in uptake which was 20 to 30 times greater for americium than for plutonium. Cline stated further that studies using plutonium should fully characterize any accompanying americium contamination before valid conclusions can be drawn.

Price (1972) conducted one of the few studies relevant to radioactive waste management involving americium. He selected Russian thistle (*Salsola kali*) and cheatgrass (*Bromus tectorum*) for investigation because of their abundance and because they are both prolific invaders of disturbed areas. The seeds of both species were germinated under controlled environmental conditions in local soil having a pH of 7.8 and spiked with $^{241}Am(NO_3)_3$. His data showed that the uptake and accumulation of americium by the Russian thistle was over twice the amount taken up by the cheatgrass. The magnitude of uptake based on the amount of americium in the rooting media was 0.032% for the Russian thistle and 0.0071% for the cheatgrass. The reason for this large difference is unknown. However, it does indicate that a degree of uncertainty exists in respect to the uptake of americium by different plant species. This difference in plant species uptake was again shown by Price (1973b and 1973c). He spiked the following organic acids, acetate as $NH_4C_2H_3O_2$, glycolate as $NaC_2H_3O_3$, oxalate as $(NH_4)_2C_2O_4$, and citrate as $(NH_4)_2HC_5H_5O_7$ with approximately 2.5 μCi/ml of ^{241}Am. Following the addition of 10 ml of each of these solutions to local soils, the seeds of Russian thistle and cheatgrass were planted and grown for approximately 2 months.

After sample analysis, he reported that a 10-fold difference frequently occurred between the amount of americium taken up by each of these species. The minimum amount of americium uptake in both species occurred in the glycolate treatment with a relative percentage uptake of 8% in the cheatgrass and 64% in the Russian thistle. The maximum uptake of americium by the Russian thistle was observed during the oxalate treatment with a relative percentage of 79%; whereas a high of 26% occurred in the cheatgrass following the acetate treatment. Also, his data showed that the apparent complexing of these organic acids in the soil resulted in a decrease in uptake when compared to his previous experiment using $^{241}Am(NO_3)_3$.

Another area in need of scientific investigation includes the identification and description of the fate and behavior of americium in both fresh water and marine ecosystems. Our knowledge of this contaminant and its distribution in these environments is limited. This is unfortunate in view of the many sources of aquatic contamination. Aside from the contribution of atmospheric deposition and wastes from reprocessing facilities (Templeton and Preston 1966), there have been more than 50 underwater and water surface nuclear tests conducted on and in the vicinity of the Eniwetok and Bikini atolls. It is perhaps easily assumed that radionuclides may be far more mobile in aquatic systems than they are in terrestrial systems. As a result, the rate of biotic incorporation of radionuclides in aquatic environments is probably enhanced. The rates and magnitude of americium incorporation into the aquatic flora is, unfortunately, unknown; however, Noshkin (1972) reported that studies are presently being conducted by several laboratories to document the concentrations and distributions of actinides in major bodies of water.

It appears that some marine plants could possibly be used as environmental indicators of selected actinides. For example, the free-floating marine algae, *Sargassum sp.*, has been reported to have a high affinity for plutonium-239. This plant has also been reported by Burkholder (1963) and Simek *et al.* (1967), to effectively concentrate other radionuclides such as cesium-137, cerium-144, praseodymium-144 and manganese-59. Recently, Cushing and Watson (1974) reported that americium-241 was found to be incorporated into the tissues of algae, and other aquatic species including several species of pond weed, *Potamogeton spp.*, following a radiological survey of a low-level radioactive waste pond.

SUMMARY

The relatively small amount of knowledge that has been gained concerning the behavior of americium in the soil-plant systems should be, at best, used conservatively in evaluating the impact of this element in our environment. Previous investigations have made it apparent that changes in pH and the addition of various concentrations of complexing or chelating agents to soils will influence the biological availability of this element in these systems.

It is easily concluded that the chelation of this radionuclide may increase plant uptake and thus lead to man via his food chain. This is a good possibility not only because of naturally occurring complexing agents, but also because of the use of commercially available chelating agents in agricultural areas to enhance the uptake and use of micronutrients by crop plants. Therefore, mechanisms of chelation should be explored.

Evidence in the literature indicates that americium can be toxic to plants and can retard normal plant growth. These effects should be explored. Also, the factors affecting the biological availability and the mobility of americium in soil systems, such as by local soil properties and microbial metabolism, need further investigation. Additionally, the need to identify the mode, rate, chemical form, and amount of americium incorporated in various plant organs is evident.

It is generally agreed that americium may become a greater environmental

liability than the most investigated transuranic element, plutonium. As a result, a complete documentation of its distribution, fate, and behavior in our environment is essential, especially in view of our expanding nuclear industry.

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